

# Online Research @ Cardiff

This is an Open Access document downloaded from ORCA, Cardiff University's institutional repository: <https://orca.cardiff.ac.uk/id/eprint/108343/>

This is the author's version of a work that was submitted to / accepted for publication.

Citation for final published version:

Kim, Minkyung, Oh, Sang Soon ORCID: <https://orcid.org/0000-0003-3093-7016>, Hess, Ortwin and Rho, Junsuk 2018. Frequency-domain modelling of gain in pump-probe experiment by an inhomogeneous medium. Journal of Physics: Condensed Matter 30 (6) , 064003. 10.1088/1361-648X/aaa473 file

Publishers page: <http://dx.doi.org/10.1088/1361-648X/aaa473>  
<<http://dx.doi.org/10.1088/1361-648X/aaa473>>

Please note:

Changes made as a result of publishing processes such as copy-editing, formatting and page numbers may not be reflected in this version. For the definitive version of this publication, please refer to the published source. You are advised to consult the publisher's version if you wish to cite this paper.

This version is being made available in accordance with publisher policies.

See

<http://orca.cf.ac.uk/policies.html> for usage policies. Copyright and moral rights for publications made available in ORCA are retained by the copyright holders.



# Frequency-domain modelling of gain in pump-probe experiment by an inhomogeneous medium

Minkyung Kim<sup>1</sup>, Sang Soon Oh<sup>2,3\*</sup>, Ortwin Hess<sup>3</sup> and Junsuk Rho<sup>1,4†</sup>

<sup>1</sup>Department of Mechanical Engineering, Pohang University of Science and Technology (POSTECH), Pohang 37673, Republic of Korea

<sup>2</sup>School of Physics and Astronomy, Cardiff University, Cardiff CF24 3AA, United Kingdom

<sup>3</sup>The Blackett Laboratory, Department of Physics, Imperial College London, London SW7 2AZ, United Kingdom

<sup>4</sup>Department of Chemical Engineering, Pohang University of Science and Technology (POSTECH), Pohang 37673, Republic of Korea

E-mail: \*[ohs2@cardiff.ac.uk](mailto:ohs2@cardiff.ac.uk) and †[jsrho@postech.ac.kr](mailto:jsrho@postech.ac.kr)

**Abstract:** Introduction of a gain medium in lossy plasmonic metamaterials reduces and compensates losses or even amplifies an incident light often with nonlinear optical effect. Here, optical gain in a pump-probe experimental setup is effectively calculated in the frequency-domain by approximating a gain material as an inhomogeneous medium. Spatially varying local field amplitudes of the pump and probe beams are included in the model to reproduce the inhomogeneous gain effect, in which population inversion occurs most strongly near the surface and decays along the propagation direction. We demonstrate that transmission spectra calculated by this method agree well with finite-difference time-domain (FDTD) simulation results. This simplified approach of gain modelling offers an easy and reliable way to analyze wave propagation in a gain medium without nonlinear time-domain calculation.

## 1. Introduction

Research on artificially structured materials with exotic optical behavior, called metamaterials, has made immense progress for several decades enabling negative refractive index [1, 2], huge artificial magnetism [3] and super-resolution imaging [4, 5]. Being composed of subwavelength dimensions, metamaterials are usually described as a homogeneous medium with effective optical parameters such as effective permittivity and permeability despite the spatially non-uniform artificial nanostructures [6]. However, resonances are generally accompanied in their optical responses by extraordinary optical parameters such as negative or extremely high refractive index, which results in high optical losses. Since the losses hinder realization and practical application of metamaterials, a new mechanism to overcome the intrinsic limitations are in demand. Gain materials, including fluorescent dye and semiconductors, which absorb incident light and emit light with lower energy, have been reported to provide field amplification to compensate the losses [7]. The inclusion of such gain materials reduces the optical losses of metamaterials or even enables metamaterials with negative losses, in other words, amplification [8-10].

Gain materials are usually modelled as a four-level system [10-13], or they can be simply described as a homogeneous medium with effective gain permittivity [14, 15]. However, since the excitation of ground-level electrons to a higher level by the photon absorption occurs locally rather than globally, the gain effect is inherently not uniform throughout the whole gain medium. Instead, the attenuation of pump beam leads to a spatially non-uniform gain effect which makes the gain stronger near the front surface and degraded as the beam propagates. In this paper, we propose a new modelling approach of gain medium where an inhomogeneous gain effect is taken into account. Refractive index as a function of propagation distance and transfer matrix method (TMM) of electromagnetic fields in a multilayer system are combined to offer frequency-domain calculations of the gain in a four-level system. Our model consists of two steps. First, the interaction between a gain medium and the pump beam generates spatially non-uniform population difference. Then, for the given population difference profile, absorption and stimulated emission caused by the propagation of probe are simulated. Electric

field amplitudes, population inversions and transmission spectra for a finite-thickness slab of an artificial gain material will be calculated and compared using (i) standard nonlinear four-level one-electron system, (ii) homogeneous gain and (iii) inhomogeneous gain approach. This approach will enable a simplified but relatively accurate calculation of wave propagation in gain materials.

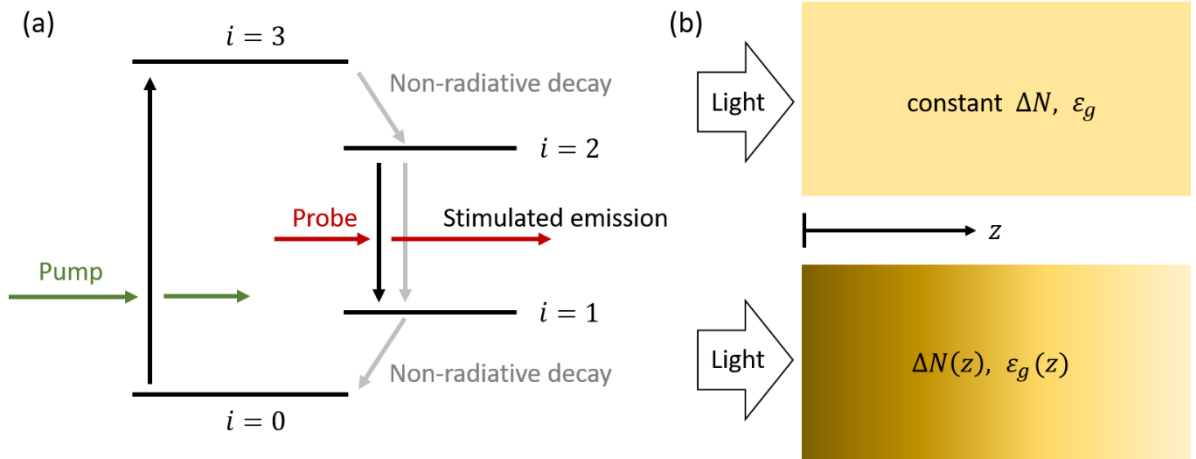
## 2. Effective pump-probe experiment in a frequency-domain

In this paper, we will focus on pump-probe experiments where a high-intensity pump beam with narrow bandwidth excites the gain medium first, and then relatively weak and broadband probe beam is used to detect the nonlinear effects with a time delay  $\Delta t_{\text{delay}}$  after the pump beam has been turned off. In the visible wavelength range, for example, the pump and the probe beam correspond to picosecond laser and white light source with femtosecond scale pulse length respectively, and the  $\Delta t_{\text{delay}}$  is on the order of tens of picosecond. To model the gain effect, we consider a four-level one-electron system (Figure 1(a)) as it successfully describes gain effect in dye materials. Especially, Rhodamine 800 (refractive index  $n = 1.62$ , total carrier density  $\bar{N} = 6 \times 10^{24} \text{ m}^{-3}$ ) with an area  $A$  is pumped by 4 picosecond pulse laser ( $\Delta t_{\text{pulselength}} = 4 \times 10^{-12} \text{ s}$ ) with an amplitude of  $E_0 = 2 \times 10^7 \text{ V/m}$ .

### 2.1 Local field effect in a gain medium

For a slab of a single uniform gain medium, the inhomogeneous gain effect originates from two factors, the attenuation of pump beam along the propagation direction and the oscillatory field intensity due to the interference between the forward and backward waves. The former results in non-uniform gain feature in a scale longer than the pump wavelength and the latter in the order of a half of the pump wavelength. Firstly, to ensure the optical pump generates inhomogeneous population inversion without reaching the saturation regime, the pump energy should be comparable to the maximum absorbable energy. Here, the maximum absorbable energy is the photon energy required to

excite all electrons in the lower level ( $i = 0$ ) to the higher level ( $i = 3$ ) during propagation of one wavelength:  $E_{\text{abs}} = A\lambda_a \bar{N} \hbar \omega_a / n$ , where  $\lambda_a$  and  $\omega_a$  are the absorption wavelength and the absorption angular frequency respectively. The pump beam energy is the incident power multiplied by the pulse length:  $E_{\text{pump}} = nc_0 \epsilon_0 A E_0^2 \Delta t_{\text{pulselength}} / 2$  where  $\epsilon_0$  is the vacuum permittivity and  $c_0$  is the light speed in free space. For the parameters chosen for our pump-probe experiment (see above), the maximum absorbable energy ( $E_{\text{abs}} = 0.74$  J) is not negligible compared with the pump energy ( $E_{\text{pump}} = 3.44$  J). It implies that the pump loses a considerable amount of its energy by exciting the lower level electrons to the higher states when it enters the slab and the attenuated pump beam excites fewer electrons as it propagates through the slab. In addition, since the electric field at a given point is the sum of forward and backward travelling waves, it will show an oscillatory spatial profile rather than a modest change in amplitude. Therefore, a slab of gain material should be described as an inhomogeneous medium with spatially varying gain parameters as shown in Figure 1(b) instead of a uniform effective medium with constant population inversion  $\Delta N$  and gain permittivity  $\epsilon_g$ .



**Figure 1.** (a) Gain mechanism of a four-level system and (b) schematics of homogeneous gain assumption (top) and inhomogeneous gain assumption (bottom).  $\Delta N$  and  $\epsilon_g$  are the population inversion and effective permittivity due to the gain.

## 2.2 Relation between the electric field and effective gain permittivity

Our four-level system is assumed to have only two optical transitions: absorption (transition  $0 \leftrightarrow 3$ ) and stimulated emission (transition  $1 \leftrightarrow 2$ ). Three non-radiative decay processes to the right below each level are added to describe temporal dynamics of carrier densities. Time evolution of carrier densities in four-level systems follows these coupled equations [11]:

$$\begin{aligned}\frac{\partial N_3}{\partial t} &= \frac{1}{\hbar\omega_a} \left( \frac{\partial \mathbf{P}_a}{\partial t} + \frac{\Delta\omega_a}{2} \mathbf{P}_a \right) \cdot \mathbf{E}_{loc} - \frac{N_3}{\tau_{32}}, \\ \frac{\partial N_2}{\partial t} &= \frac{1}{\hbar\omega_e} \left( \frac{\partial \mathbf{P}_e}{\partial t} + \frac{\Delta\omega_e}{2} \mathbf{P}_e \right) \cdot \mathbf{E}_{loc} + \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{21}}, \\ \frac{\partial N_1}{\partial t} &= -\frac{1}{\hbar\omega_e} \left( \frac{\partial \mathbf{P}_e}{\partial t} + \frac{\Delta\omega_e}{2} \mathbf{P}_e \right) \cdot \mathbf{E}_{loc} + \frac{N_2}{\tau_{21}} - \frac{N_1}{\tau_{10}}, \\ \frac{\partial N_0}{\partial t} &= -\frac{1}{\hbar\omega_a} \left( \frac{\partial \mathbf{P}_a}{\partial t} + \frac{\Delta\omega_a}{2} \mathbf{P}_a \right) \cdot \mathbf{E}_{loc} + \frac{N_1}{\tau_{10}}.\end{aligned}\tag{1}$$

Here, the subscript  $a$  and  $e$  indicate absorption and emission respectively.  $\omega_a$  and  $\omega_e$  are the resonance angular frequencies,  $\Delta\omega_a$  and  $\Delta\omega_e$  are the full bandwidths,  $\mathbf{P}_a$  and  $\mathbf{P}_e$  are the polarization densities for absorption and emission respectively.  $N_i$  is carrier density in the  $i$ -th level ( $i = 0, 1, 2, 3$ ) and the total carrier density  $\bar{N}$  is given by  $\bar{N} = N_0 + N_1 + N_2 + N_3$ .  $\tau_{ij}$  is the lifetime of carriers for the non-radiative decay transition from  $i$  to  $j$ . Other non-radiative terms such as successive decays are assumed to be negligible.  $\mathbf{E}_{loc}$  is a local electric field given by  $[(\epsilon + 2)/3]\mathbf{E}$  from the Lorentz approximation, not the average electric field [16].

In equation (1), the terms that contain the polarization densities  $\mathbf{P}_i$  and local electric field  $\mathbf{E}_{loc}$  correspond to excitation of the ground-level electrons and stimulated emission for  $i = a$  and  $i = e$  respectively while the other terms represent non-radiative relaxation processes. Gain materials such as dyes or semiconductors which are good candidates to be implemented in metamaterials have long  $\tau_{21}$  compared to  $\tau_{32}$  and  $\tau_{10}$  [17], so that electrons in the third and first level rapidly decay to the lower level, but stay in the second level. The second level is sometimes called as a metastable level for its

relatively long lifetime [12]. In other words, electrons in the first and the third level relax fast whereas they stay in the second level longer compared to the time delay ( $\tau_{10}, \tau_{32} \ll \Delta t_{\text{delay}} \ll \tau_{21}$ ). Therefore, a probe beam experiences the gain materials in which the electrons relaxed from the highest level stay in the metastable level giving rise to a population inversion.

To calculate the absorption and gain effect, population difference of two optical transitions  $0 \leftrightarrow 3$  and  $1 \leftrightarrow 2$  should be known. In the remaining part of this section, we will find a formula of the spatial-dependent population difference and then show that effective permittivity for the pump and the probe beam can be expressed in terms of electric field.

For a ground state before the excitation, the initial condition is given by  $N_0 = \bar{N}$ ,  $N_i = 0$  ( $i = 1, 2, 3$ ). As a pulse of the pump beam propagates in the gain medium, the carrier densities vary in time and then after the pump is turned off and before the probe is turned on, they reach a state which is different from the initial state. Since the carrier densities in all level vary slowly due to the scale mismatch of the lifetime and time delay between the pump and the probe, we approximate this state as a stationary state. In the absence of the probe beam, the carrier density in each level at the stationary state can be obtained by applying  $\partial/\partial t = 0$  to the second and third equations of (1). Then the carrier densities for  $i = 1, 2, 3$  at the stationary state follow

$$N_1:N_2:N_3 = \tau_{10}:\tau_{21}:\tau_{32} \quad (2)$$

Additionally, the time evolution of  $N_3$  can be also expressed as a simplified form in terms of stimulated transition probability  $\Gamma_{ij}$  [12].

$$\frac{\partial N_3}{\partial t} = \Gamma_{03}N_0 - \frac{N_3}{\tau_{32}} \quad (3)$$

We will now derive the expression for the spatial-dependent  $\Gamma_{03}$  starting from a formula  $\Gamma_{03} = \sigma_{abs}I/\hbar\omega_a$  [14] where  $I(z, t) = c_0 n(z) \epsilon_0 |E(z, t)|^2/2$  is a local intensity and  $\sigma_{abs}(z) = (\lambda_a/n)^2 \gamma_{rad}(z)/2\pi\Delta\omega_a$  is the absorption cross section. The Lorentzian term in the absorption cross section in Ref. [14] can be neglected by assuming the narrow bandwidth of the pump beam. The

radiative decay rate  $\gamma_{rad}$  is given by Fermi's golden rule [18]:  $\gamma_{rad}(z) = e^2 d_a^2 \omega_a^3 / \pi \epsilon \hbar c^3$ . Here,  $d_i$  is dipole length when the dipole is oriented parallel to the incident polarization. Note that  $\epsilon$  and  $c$  are the permittivity and the speed of light in the medium, both of which depend on the propagation distance  $z$ . In other words, the absorption cross section and the intensity are both functions of propagation distance. However,  $\Gamma_{03}(z, t) = e^2 d_a^2 |E(z, t)|^2 / \hbar^2 \Delta \omega_a$  is dependent only on the field amplitude, but not on refractive index as  $n(z)$  are cancelled out. Therefore, spatially varying population difference is explicitly determined by the local field amplitude. Note that the population difference is only dependent on the wave propagation direction but uniform with respect to the wavefront plane due to plane symmetry.

We introduce a time-independent variable  $\Gamma_{03,eff}$  to effectively describe the stimulated transition probability in the frequency domain using slowly varying envelope approximation. The electric field of pump has the Gaussian envelope with an oscillating term of its angular frequency:  $E(z, t) = E(z) \sin(-\omega_c(t - t_0)) \exp(-(t - t_0)^2 / 2\Delta t^2)$  where  $t_0$  is center of the Gaussian envelope and  $\Delta t = \Delta t_{pulselength} / 2\sqrt{\log 2}$ . First, ignoring the fast oscillating term  $\sin(-\omega_c(t - t_0))$ , a crude approximation will be taking average of the Gaussian envelope in time, which leads to effective electric field amplitude as  $E_{0,eff}(z) = E_0(z) \sqrt{2\pi} \Delta t / \Delta t_{delay}$ . Then, we can obtain  $\Gamma_{03,eff}$  by substituting  $E_{0,eff}(z)$  instead of  $E(z, t)$  into  $\Gamma_{03}(z, t)$  and multiplying additional  $1/2$  to take into account the square term of  $\sin(-\omega_c(t - t_0))$ . To sum up,  $\Gamma_{03,eff} = (\sqrt{\pi} \Delta t / \Delta t_{delay})^2 e^2 d_a^2 |E(z)|^2 / \hbar^2 \Delta \omega_a$  is used instead of  $\Gamma_{30}$  to remove time dependence. Finally, by incorporating equation (2) with (3), we obtained the expression for the population difference of both absorption and emission which are defined as  $\Delta N_a = N_3 - N_0$  and  $\Delta N_e = N_2 - N_1$ :

$$\Delta N_a^{stat} = N_3 - N_0 = \frac{\tau_{32} \Gamma_{03,eff} - 1}{1 + (\tau_{32} + \tau_{21} + \tau_{10}) \Gamma_{03,eff}} \bar{N}, \quad (4)$$

$$\Delta N_e^{stat} = N_2 - N_1 = \frac{(\tau_{21} - \tau_{10}) \Gamma_{03,eff}}{1 + (\tau_{32} + \tau_{21} + \tau_{10}) \Gamma_{03,eff}} \bar{N}. \quad (5)$$



Meanwhile, the population densities are governed by the following equations of motion for  $i = a$  and  $i = e$  [14].

$$\frac{\partial^2 \mathbf{P}_i}{\partial t^2} + \Delta\omega_i \frac{\partial \mathbf{P}_i}{\partial t} + \omega_i^2 \mathbf{P}_i = -\sigma_i \Delta N_i \mathbf{E}_{loc} \quad (6)$$

The coupling constant  $\sigma_i$  of  $\mathbf{P}_i$  to the local electric field is provided by  $\sigma_i = 2\omega_i e^2 d_i^2 / \hbar$  [11]. The coupling constant and the population difference determine the strength of the coupling between the induced polarization density and the local electric field and thereby effective gain permittivity. Given that pump beam usually has a picosecond scale pulse length for a narrow bandwidth and the relatively broadband probe is in tens of femtoseconds scale for visible range, both pump and probe beams can be considered as continuous waves for their relatively long pulse lengths compared to the wavelength. Therefore, we can simplify the partial differential equations to get effective permittivity using the time-harmonic assumption.

$$\begin{aligned} \mathbf{D}_i &= \varepsilon_0 \mathbf{E} + \mathbf{P}_i = \varepsilon_0 \varepsilon \mathbf{E} \\ \varepsilon &= \varepsilon_r + \varepsilon_g = \varepsilon_r + \frac{1}{\varepsilon_0} \frac{\sigma_i \Delta N_i}{\omega^2 + i\omega\Delta\omega_i - \omega_i^2} \end{aligned} \quad (7)$$

The total relative permittivity  $\varepsilon$  is a sum of relative permittivity  $\varepsilon_r$  of the gain material itself and effective gain permittivity  $\varepsilon_g$  with Lorentzian line shape.

To calculate the changes of the population difference and permittivity in the gain medium, let us consider the propagation of the pump beam which corresponds to absorption process by exciting electrons in the lowest level to the higher levels. Strictly speaking, the excitation of electrons by the pump is a transient process, which requires nonlinear time-domain simulation. In other words, the effective gain permittivity is a function of both time and propagation distance. To remove the time dependence, we used population difference for absorption by averaging the population difference of the initial state ( $\Delta N_a = -\bar{N}$ ) and that of the stationary state ( $N_a^{\text{stat}}$ ). Then, we can effectively describe the interaction between the pump and the gain slab as a wave propagation in an inhomogeneous medium with the following effective gain permittivity.

$$\varepsilon_g^{\text{pump}} = \frac{1}{\varepsilon_0} \frac{\sigma_a(-\bar{N} + \Delta N_a^{\text{stat}})/2}{\omega^2 + i\omega\Delta\omega_a - \omega_a^2} \quad (8)$$

Here, the negative sign of the population difference indicates a loss. Note that the contribution of optical transition  $2 \leftrightarrow 1$  can be disregarded here as the pump beam does not overlap with emission wavelength. On the other hand, the propagation of probe beam is a more complicated process which involves both absorption and stimulated emission. The probe beam usually has broad wavelength range centered at the emission wavelength. In other words, the probe contains waves with absorption wavelength as well as those with emission wavelength. The wave with absorption wavelength works as if it is another pump during the propagation of the gain medium. It excites the electrons in  $i = 0$  level to  $i = 3$  level and decays. In contrast, the wave with emission wavelength induces radiative emission of electrons in  $i = 2$  level to  $i = 1$  level. Therefore, the probe beam experiences a medium with two Lorentzian permittivities, one is positive (loss from absorption) and the other is negative (gain from stimulated emission).

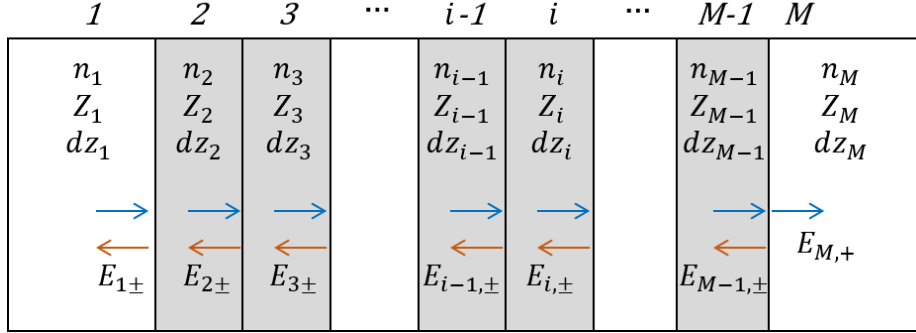
$$\varepsilon_g^{\text{probe}} = \frac{1}{\varepsilon_0} \frac{\sigma_a \Delta N_a^{\text{stat}}}{\omega^2 + i\omega\Delta\omega_a - \omega_a^2} + \frac{1}{\varepsilon_0} \frac{\sigma_e \Delta N_e^{\text{stat}}}{\omega^2 + i\omega\Delta\omega_e - \omega_e^2} \quad (9)$$

Here, the spatially non-uniform parameters in effective gain permittivity of both pump and probe are population difference  $N_a^{\text{stat}}$  and  $N_e^{\text{stat}}$ , which can be easily obtained by the local field amplitude as stated earlier. All the other parameters are constant. Therefore, we can get effective permittivity if the field amplitude is known. Importantly, the change of the carrier densities caused by the probe can be neglected as the intensity of the probe beam is several orders of magnitude smaller than that of the pump beam.

So far, a frequency-domain approach has been introduced for an effective modelling of the gain medium in a pump-probe experiment with narrowband laser pump and broadband probe beams. Based on stationary state assumption, time-harmonic assumption and slowly varying envelope approximation, we proposed an approach to calculate effective gain permittivity in frequency-domain.

In the next section, we will discuss how to describe the propagation of the pump and probe beams through the spatially varying effective medium.

### 2.3 Transfer matrix method for an inhomogeneous medium



**Figure 2.** Wave propagation in a medium of spatially varying optical parameters.

To calculate the electromagnetic fields in the inhomogeneous medium, we employ the transfer matrix method for a dielectric multilayer film [19, 20]. Wave propagation in a medium with spatially varying permittivity can be readily calculated by taking a limit of thickness to zero in the dielectric multilayer problem. Suppose a dielectric multilayer slab is placed between two semi-infinite media labelled as  $i = 1$  and  $i = M$ . We assume that the dielectric media are non-magnetic ( $\mu = 1$ ). The slab is composed of  $M - 2$  layers ( $i = 2, 3, \dots, M - 1$ ), where  $i$ -th layer has refractive index  $n_i$ , impedance  $Z_i$  and thickness  $dz_i$ , and the total gain slab has thickness  $L = \sum_{i=2}^{M-1} dz_i$ . Then the electric and magnetic fields at the layer  $i - 1$  and the layer  $i$  are related by transfer matrix  $T_i$  according to continuity equation [20]:

$$\begin{pmatrix} E_{i-1} \\ H_{i-1} \end{pmatrix} = T_i \begin{pmatrix} E_i \\ H_i \end{pmatrix} = \begin{pmatrix} \cos n_i k_0 dz_i & -jZ_i \sin n_i k_0 dz_i \\ -jZ_i^{-1} \sin n_i k_0 dz_i & \cos n_i k_0 dz_i \end{pmatrix} \begin{pmatrix} E_i \\ H_i \end{pmatrix} \quad (10)$$

for  $i = 1, 2, \dots, M$  with  $dz_M = 0$  and the initial condition  $\begin{pmatrix} E_M \\ H_M \end{pmatrix} = \begin{pmatrix} 1 \\ 1/Z_i \end{pmatrix} E_M$ . Taking a limit of  $dz_i \rightarrow 0$  for  $i = 2, 3, \dots, M - 1$  gives the first order differential equation with zero diagonal terms. It

is solvable for a constant or simple form of refractive index. However, since the refractive index of each layer is dependent on the electric field amplitude as described in the previous section, wave propagation in the inhomogeneous gain medium requires a TMM calculation with an infinitesimal thickness. If  $\begin{pmatrix} E_i \\ H_i \end{pmatrix}$  is known,  $n_i$  can be calculated by the equation (8) and equation (9) for pump and probe beams respectively. Then  $\begin{pmatrix} E_{i-1} \\ H_{i-1} \end{pmatrix}$  can be obtained from the equation (10). Consequently, the electromagnetic fields in the layer  $i$  is given by

$$\begin{pmatrix} E_i \\ H_i \end{pmatrix} = \left( \prod_{l=i+1}^M T_l \right) \begin{pmatrix} E_M \\ H_M \end{pmatrix}. \quad (11)$$

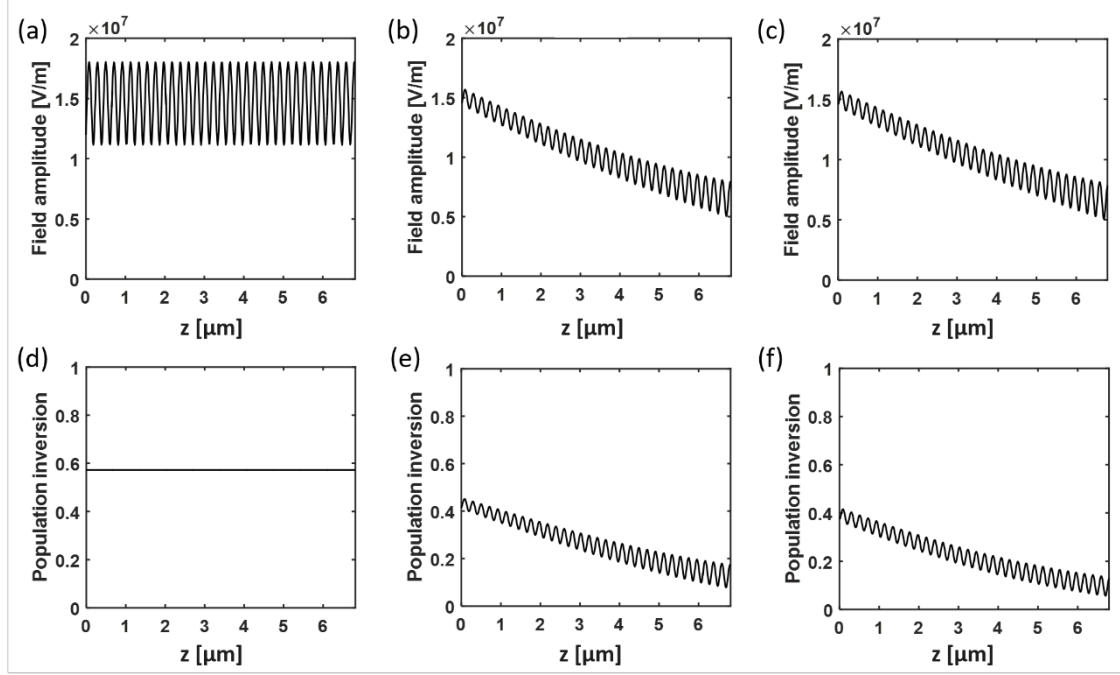
Combined with equation (8) and (9), the equation (11) fully determines the electromagnetic fields at every dielectric layer. Furthermore, we can get transmission and reflection spectra by  $E_{M+}/E_{1+}$  and  $E_{1-}/E_{1+}$  respectively.

#### 2.4 Numerical simulations and comparison

We performed a numerical pump-probe experiment with a gain slab using (i) nonlinear calculation in time-domain, (ii) homogeneous gain and (iii) inhomogeneous gain approach. Rhodamine 800 of thickness  $6.8 \mu\text{m}$ , which corresponds to ten times of the pump wavelength, is used and modelled as 500 layers with an equal thickness. Since the overall thickness is comparable to the wavelength, the slab acts as a Fabry-Perot resonator with oscillatory transmission spectrum. Therefore, the thickness is determined to have a unity transmission at the emission wavelength to exclude the effect from multiple reflections. We also assume the gain material to have zero imaginary part of the refractive index in the absence of pump to distinguish gain effect from the attenuation from inherent losses.

The simulation parameters are chosen as follows [11] :  $\sqrt{\epsilon_r} = 1.62$ ,  $\lambda_a = 680 \text{ nm}$ ,  $\lambda_e = 710 \text{ nm}$ ,  $\Delta\omega_a = \Delta\omega_e = 1 \times 10^{14} \text{ s}^{-1}$ ,  $\tau_{32} = \tau_{10} = 1 \times 10^{-13} \text{ s}$ ,  $\tau_{30} = \tau_{21} = 5 \times 10^{-10} \text{ s}$ ,  $d_a = 0.1 \text{ nm}$ ,  $d_e = 0.09 \text{ nm}$  and  $\bar{N} = 6 \times 10^{24} \text{ m}^{-3}$ . The incident pump amplitude is set as  $2 \times 10^7 \text{ V/m}$ . For

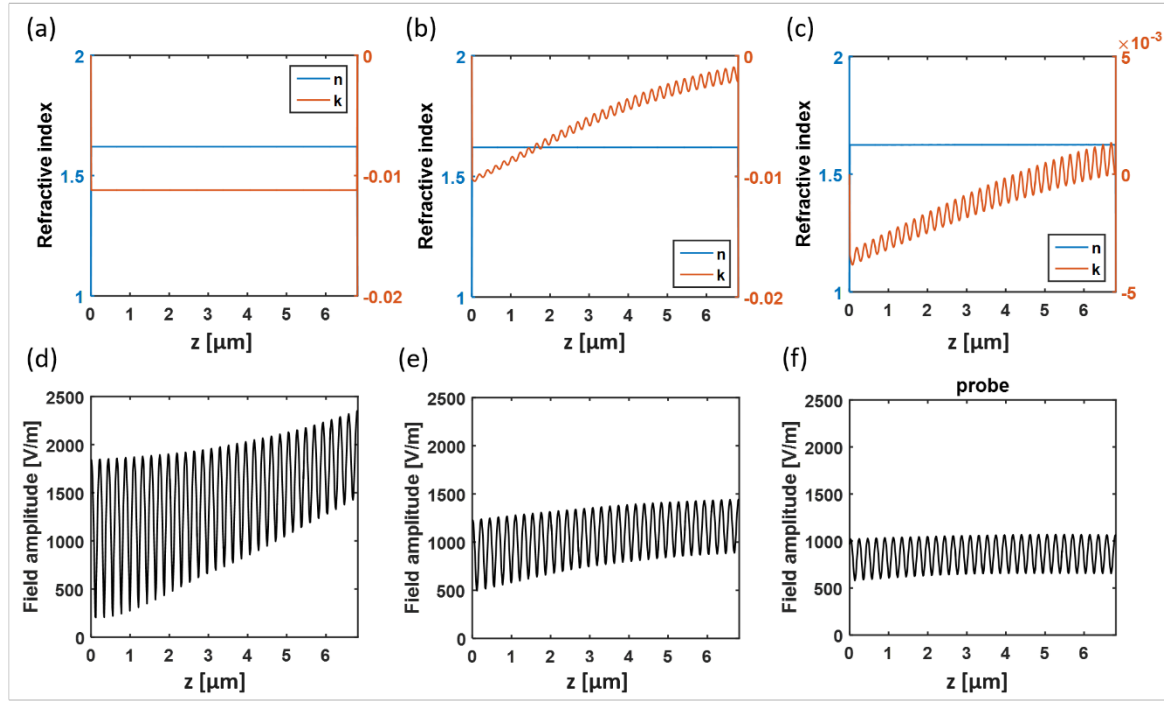
time-domain calculations, we used Lumerical FDTD solution [21] with an additional plugin. In the FDTD simulation, the pulse lengths of pump and probe beams are set to  $4 \times 10^{-12}$  s and  $1.2 \times 10^{-14}$  s respectively with  $2.5 \times 10^{-11}$  s delay between the pump and probe.



**Figure 3.** (a)-(c) Pump amplitude and (d)-(f) population inversion  $N_2 - N_1$  normalized by the total carrier density, calculated by homogeneous gain approach (left column), inhomogeneous gain approach (middle column) and FDTD (right column), respectively.

Electric fields calculated by the homogeneous gain analysis, inhomogeneous gain approach and the FDTD are shown in Figure 3 (a)-(c) respectively. In the homogeneous assumption, the field amplitude of pump exhibits standing wave-like amplitude originated from the interference of waves propagating forward (transmitting) and backward (reflecting) direction as shown in Figure 3(a). Instead of the oscillatory amplitude, incident field amplitude is used to calculate population inversion, which leads to constant population inversion which is overestimated throughout the slab (Figure 3(d)). On the contrary, local field effect included in inhomogeneous gain analysis gives rise to distinct

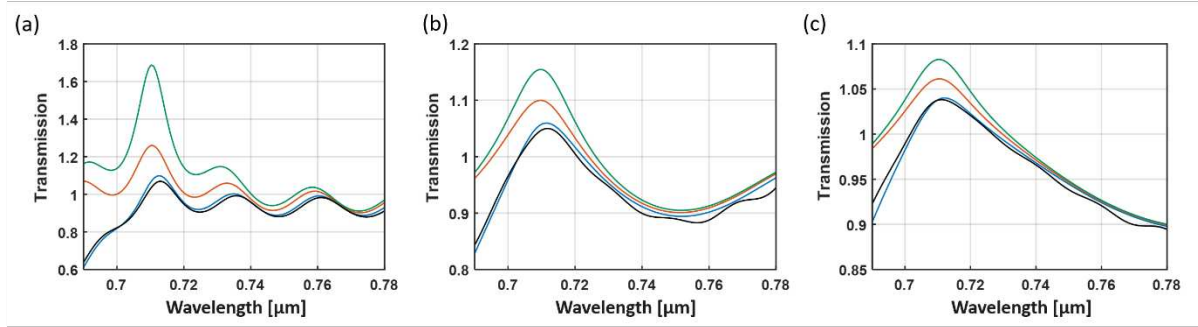
characteristics. The pump attenuates as well as oscillates during the propagation as shown in Figure 3(b). Affected by the local field amplitude, the population inversion also decays with oscillations during the propagation which agrees well with the FDTD calculation results (Figure 3(f)).



**Figure 4.** (a)-(c) Refractive index at emission wavelength where  $n$  and  $k$  are real and imaginary part, and (d)-(f) probe amplitude, calculated by homogeneous gain approach (left column), by inhomogeneous gain approach while neglecting (middle column) and including (right column) the absorption of probe beam, respectively.

Due to the modulated population inversion, real and imaginary part of the refractive index ( $n + ik$ ) at emission wavelength varies in space for the inhomogeneous gain model and FDTD whereas the refractive index is constant for the homogeneous gain model (Figure 4(a) to 4(c)). Since the amplitude of the incident probe is much smaller than that of the pump, we simulated two cases where the absorption of probe, which corresponds to the first term in the right-hand side of equation (9), is

neglected (middle column) and included (right column). In both cases, the imaginary part of the refractive index increases implying the diminished gain effect during the propagation. When the absorption is not included, the imaginary part of the refractive index is negative over the entire slab meaning that the probe beam will experience only the gain effect. Accordingly, the electric field amplifies gradually despite the negative gradient. On the other hand, if the absorption is included, the imaginary part of the refractive index becomes positive after some point and the electric field starts to decay. It means that even at the emission wavelength, absorption occurs and even exceeds the stimulated emission after several wavelengths.



**Figure 5.** Transmission spectra calculated by homogeneous gain approach (green), inhomogeneous gain approach by neglecting the probe absorption (red), inhomogeneous gain approach by considering the probe absorption (blue) and FDTD (black), respectively. The thickness of slab is (a) 6.8 μm, (b) 1.97 μm and (c) 1.1 μm.

To check the functionality of our inhomogeneous gain approach, the transmission spectra of the probe beam are calculated with the homogeneous gain analysis, inhomogeneous gain approach and the FDTD for three different slab thickness: 6.8 μm, 1.97 μm and 1.1 μm (Figure 5). These thicknesses are chosen to give unity transmission without gain, but the scheme is not limited to this condition and applicable for an arbitrary thickness. Transmission spectra obtained from homogeneous gain approach is indicated by green lines with an oscillating feature due to the multiple reflections.

Here, the gain effect is overestimated especially when the slab is several orders thicker than the pump wavelength. Although the inhomogeneous gain approach with neglected probe absorption shows reduced gain (red), which is similar to FDTD calculation (black), it fails to explain low transmission near the absorption wavelength originating from the excitation of electrons by the probe beam. However, if the probe absorption is included (blue), the calculated transmission contains both the gain at emission wavelength and the loss at absorption wavelength. A notable feature here is that the inhomogeneous gain calculated in frequency-domain perfectly reproduces transmission spectra simulated by nonlinear time-domain calculation. A numerical pump-probe experiment which has been regarded to be available at only time-domain can be performed in the frequency-domain either for this simple slab geometry.

### **3. Conclusion**

In conclusion, optical gain in a pump-probe experiment is calculated on the basis of an inhomogeneous gain approach and compared with a finite-difference time-domain (FDTD) analysis and the usual approach assuming a spatially homogeneous gain distribution. By considering absorption of the incident photons' energy, we include the local field distribution into the effective gain approach, allowing us to relax the constant field assumption. Successful reproduction of oscillatory and decaying shape of the pump and probe beam is presented. We also demonstrate that the inhomogeneous gain approach can be readily used to calculate transmission spectra of a probe beam for a slab with a finite thickness, which shows excellent agreement with the FDTD results. The current study can generally be applied to complex three-dimensional gain geometries, but for comparison with the transmission matrix method is here restricted to one-dimensional slabs. We anticipate that the simplified analysis for gain materials can also be performed for loss-compensated metamaterials for real applications, providing speedy and accurate modelling of gain materials such as dye or semiconductor.



## **Acknowledgments**

This work is financially supported by the National Research Foundation grants (NRF-2017R1E1A1A03070501, NRF-2015R1A5A1037668, CAMM-2014M3A6B3063708) funded by the Ministry of Science, ICT and Future Planning (MSIP) of the Korean government and part-funded by the European Regional Development Fund through the Welsh Government. M.K. acknowledges Global Ph.D. Fellowships (NRF-2017H1A2A1043204) from NRF-MSIP of Korean government.

## **ORCID iDs**

Junsuk Rho <https://orcid.org/0000-0002-2179-2890>

Sang Soon Oh <https://orcid.org/0000-0003-3093-7016>

Ortwin Hess <https://orcid.org/0000-0002-6024-0677>

## References

- [1] Smith D R and Kroll N 2000 *Phys. Rev. Lett* **85** 2933
- [2] Shelby R A, Smith D R and Schultz S 2001 *Science* **292** 77
- [3] Pendry J B, Holden A J, Robbins D J and Stewart W J 1999 *IEEE Trans. Microw. Theory Techn.* **47** 2075
- [4] Rho J, Ye Z, Xiong Y, Yin X, Liu Z, Choi H, et al. 2010 *Nat. Commun.* **1** 143
- [5] Pendry J B 2000 *Phys. Rev. Lett.* **85** 3966
- [6] Pendry J B, Holden A J, Stewart W J and Youngs I 1996 *Phys. Rev. Lett.* **76** 4773
- [7] Zheludev N I, Prosvirnin S L, Papasimakidis N and Fedotov V A 2008 *Nat. Photon.* **2** 351
- [8] Hess O, Pendry J B, Maier S A, Oulton R F, Hamm J M and Tsakmakidis K L 2012 *Nat. Mater.* **11** 573
- [9] Xiao S, Drachev V P, Kildishev A V, Ni X, Chettiar U K, Yuan H K, et al. 2010 *Nature* **466** 735
- [10] Wuestner S, Pusch A, Tsakmakidis K L, Hamm J M, Hess O 2011 *Philos. Trans. A Math. Phys. Eng. Sci.* **369** 3525
- [11] Wuestner S, Pusch A, Tsakmakidis K L, Hamm J M and Hess O 2010 *Phys. Rev. Lett.* **105** 127401
- [12] Fang A, Huang Z, Koschny T, Soukoulis C M 2011 *Opt. Express* **19** 12688
- [13] Taflove A 2006 *Opt. Express* **14** 1702
- [14] Siegman A E 1986 *Lasers* (University Science Books)
- [15] Campione S, Albani M and Capolino F 2011 *Opt. Mater. Express* **1** 1077
- [16] de Vries P and Lagendijk A 1998 *Phys. Rev. Lett.* **81** 1381
- [17] Sperber P, Spangler W, Meier B and Penzkofer A 1998 *Opt. Quantum. Electron.* **20**, 395
- [18] Henderson B and Imbusch G F 1989 *Optical spectroscopy of inorganic solids* (UK Clarendon Press)
- [19] Yeh P 2005 *Optical waves in layered media* (Wiley)
- [20] Orfanidis S J 2016 *Electromagnetic waves and antennas* (Rutgers University)
- [21] Lumerical FDTD Solution, <http://www.lumerical.com/>